



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

GROUP ART UNIT: 2826

Toshio SAKAI et al.

SERIAL NO. : 10/828,323

EXAMINER: Thomas L. Dickey

FILED : April 21, 2004

FOR : ORGANIC ELECTROLUMINESCENT DEVICE WITH
PHENYLENEDIAMINE HOLE INJECTOR AND CUPC
INTERMEDIARY

DECLARATION UNDER 37 CFR 1.132

Honorable Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

Now comes Hisayuki KAWAMURA who deposes and states that:

1. I am a graduate of Department of Science at Kyoto University and received my master's degree in March, 1991.
2. I have been employed by IDEMITSU KOSAN Co., Ltd. for 14 years as a research associate at Central Research Laboratory of IDEMITSU KOSAN Co., Ltd.
3. I am one of the named inventors of the present U.S. Patent Application.
4. I have further reviewed the cited references of Hosokawa et al. (US 5,364,654) and Imai et al. (US 5,374,489).
5. The following experiments were performed by me or under my direct supervision and control.

Object of Experiment

In order to demonstrate the superiority of the present invention, I further declare and state:

Procedures of Experiment

I provided or synthesized the typical oligomer having a phenylenediamine

structure in each invention, and prepared the electroluminescence device in accordance with Examples 1 and 4 in the present invention. Further, I prepared the electroluminescence device in accordance with Hosokawa et al. (US 5,364,654) and Imai et al. (US 5,374,489). In other words, in order to compare the properties of the electroluminescence device, similar structures of the electroluminescence device were employed except the compound in the hole injection layer as follows:

Example 1 (Exactly the same as in the Description)

In this Example 1, an organic EL element was obtained by employing the following specific element construction in the above-described mode.

Incidentally, the following layers (2) to (5) were formed by vacuum deposition.

(1) Construction of an element

1) anode: ITO

2) intermediate layer:

CuPc(copper phthalocyanine)

3) hole injection layer:

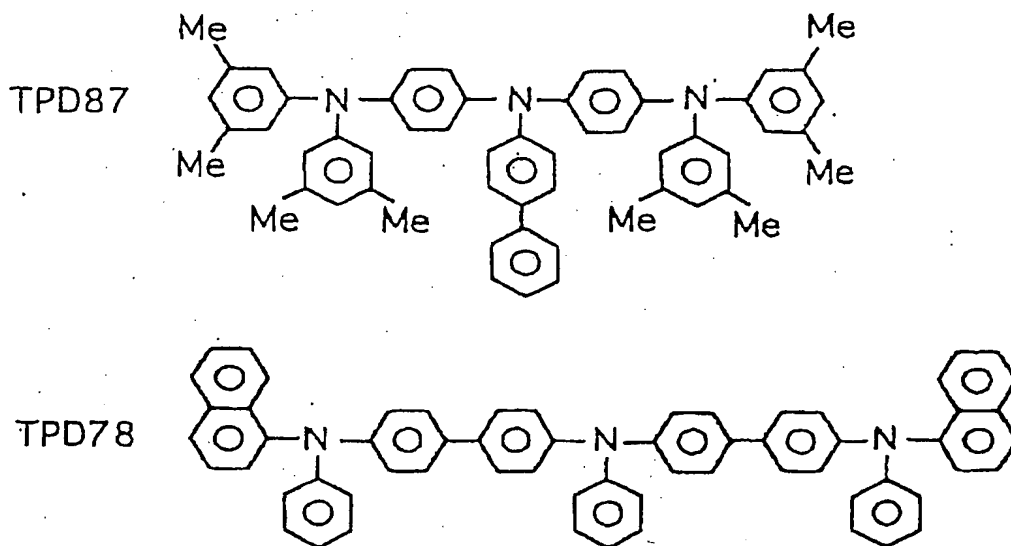
laminated film of TPD 87 and TPD 78

4) luminous layer:

Alq (tris(8-quinolinol) aluminum)

5) cathode: Composite film of Al and Li

The structural formulas of TPD 87 and TPD 78 are as follows.



Example 4

An organic EL element of this Example 4 was produced as in Example 1 using the same construction of the element as in Example 1 except that the hole injection layer was formed using TPD3 below and TPD78 instead of TPD87 and TPD78.

Comparative Example 1 (Exactly the same as in the Description)

An organic EL element of this Comparative Example 1 was obtained as in Example 1 employing the same construction of the element as in Example 1 except that the intermediate layer was omitted.

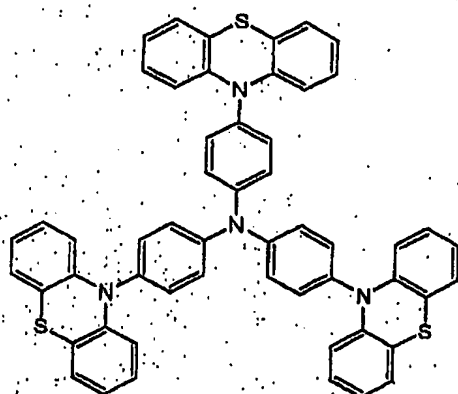
Comparative Example 5 (Corresponds to Example 1 of Imai et al.)

An organic EL element of this Comparative Example 5 was produced as in Comparative Example 1 using the same construction of the element as in Comparative Example 1 except that the hole injection layer was formed using TPD 3 below and TPD78 instead of TPD87 and TPD78. Ionized potential of TPD 3 is 5.2 eV and Tg of TPD 3 is 141 °C.

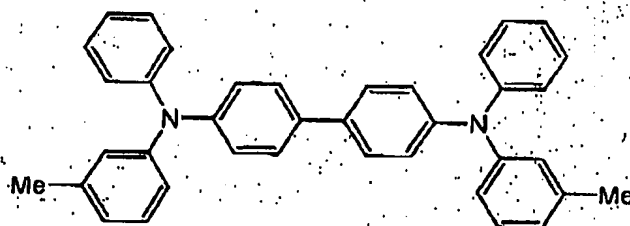
Comparative Example 6 (Corresponds to Example 18 of Hosokawa et al.)

An organic EL element of this Comparative Example 6 was produced as in Example 1 using the same construction of the element as in Example 1 except that the hole injection layer was formed using TPD4 below instead of TPD87 and TPD78; and that the luminous layer was formed using DPVBi and Alq instead of Alq only..

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TPD3



TPD4

[Evaluation of an organic EL element]

With respect to the organic EL elements of Examples 1, 4 and Comparative Examples 1, 5 and 6, the durability and the heat resistance were measured respectively.

The measurement of the durability was conducted by driving each of the organic EL elements with the initial luminance of 300 cd/m² and measuring the half-life. At this time, the driving conditions are a duty of 1/100 and a driving frequency of 60 Hz. The results are shown in Table 3.

Incidentally, in Table 3, the life of each element was shown in terms of a ratio given when the half-life in Comparative Example 1 was defined as 1.

The measurement of the heat resistance was conducted by storing each of the organic EL elements at 85°C for 500 hours and examining the change in the luminous efficiency before and after the storage. The results are shown in Table 3. In Table 3, "Good" indicated that the luminous efficiency was not decreased, and "Bad" indicated that the luminous efficiency was decreased.

Table 3

	Intermediate layer	Hole injection layer	Tg (°C)*	Life	Heat resistance
Example 1	CuPc	TPD 87/TPD 78	112/126	5	Good
Example 4	CuPc	TPD 3/TPD 78	141/126	5	Good
Comparative Example 1		TPD 87/TPD 78	112/126	1	Good
Comparative Example 5		TPD 3/TPD 78	141/126	1	Good
Comparative Example 6	CuPc	TPD 4	60	3	Bad

*Tg(°C) indicates a glass transition temperature of each material constituting a

hole injection layer.

Additionally, heat resistance of Comparative Example 6 was so bad that current efficiency degraded furiously after storing the sample for 500 hours at 85 °C because Tg of TPD 4 is 60 °C.

Consideration

As described above, it verifies that the EL devices of the present invention are superior in lifetime and heat resistance compared with prior art. We wish to point out that the compound (TPD 3 or TPD 4) in hole injecting layer of both Hosokawa et al. and Imai et al. do not satisfy the requirement of the present invention. On the other hand, Hosokawa et al. and Imai et al. exhibit inferiority in the characteristics (Comparative Example 5 and Comparative Example 6).

6. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Hisayuki Kawamura
Hisayuki KAWAMURA

Date: February 1, 2006